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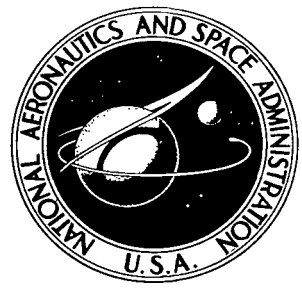
NASA TN D-4466



LIGHT-INDUCED MODULATION OF ABSORPTION (LIMA) IN CADMIUM SULFIDE

by Karl W. Böer and Edmund J. Conway

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • MARCH 1968

NASA TN D-4400

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SUMMARY

The bulk optical absorption from 0.5 micrometer to 2 micrometers of cadmium sulfide single crystals has been modulated by a chopped continuous wave laser. Data showing the intensity and chopping frequency dependences are discussed. A few of the implications of these data are also mentioned.

INTRODUCTION

A brief description is presented of some characteristics of modulated weak optical absorption in single crystals of cadmium sulfide, and an approach to a complete explanation of the data is suggested. A short genealogy of the experimental method is also presented.

Intense optical excitation can change the transmission in certain wavelength ranges and can be used for defect center analysis. Optical bleaching and an associated increase in absorption in a different wavelength range were tools employed early in the study of the defect structure and gave, for example, a very convincing indication of the connection between F and F' centers. However, this method is restricted to a few centers for which the reaction kinetics allows accumulation of redistributed charges to such an extent that they can easily be observed. The increase of excitation density by using lasers has extended the sensitivity of experimental investigations and thereby allowed observation of states with lifetimes as short as 10^{-6} second (e.g., F^* in potassium iodide) (refs. 1 and 2). With this method, an increase of absorption in cadmium sulfide near 850 nanometers has been reported recently (ref. 3). The use of modulated excitation and phase sensitive amplification of the change in optical absorption has further increased the sensitivity (ref. 4).

The authors are indebted to Hughes Research Laboratory and Spectra-Physics, Inc. for use of the krypton-ion lasers.

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EXPERIMENT, RESULTS, AND DISCUSSION

It is now possible to investigate the changes of the optical absorption caused by redistribution of electrons over a wide energy range in the band gap due to optical excitation into valence and conduction bands. This change can be accomplished by excitation with a modulated high-intensity light (chopped laser beam) and a low-intensity detection beam. This approach avoids nonlinear effects due to two-quantum absorption such as that described in reference 5. With the experimental setup sketched in figure 1, such a light induced modulation of absorption (LIMA) has been detected down to the 10^{-6} range of relative absorption.

Several cadmium sulfide crystals have been studied with this method, and all have shown LIMA in the entire extrinsic range from the fundamental edge (about 500 nanometers) up to 2 micrometers, the limiting wavelength of the optical equipment used. In general, in the range close to the absorption edge a decrease of absorption (bleaching) is observed. On the other hand, in the longer wavelength range the absorption is increased.

In figure 2, typical LIMA curves are shown for two different cadmium-sulfide crystals. ("Undoped" crystals with no known differences in preparation were used.) The magnitude of LIMA increases with increasing excitation density; however, the wavelength for which the fractional change in absorption constant $\Delta k/k$ is zero, i.e., the characteristic crossover wavelength between decreased and increased absorption, does not change markedly with excitation density. This result can be seen from curves (1) and (2) in figure 2, for which the excitation density was changed by using two different laser wavelengths 647.1 nanometers and 530.8 nanometers, the shorter wavelength flux being more strongly absorbed in the crystal (both laser beams having about the same density of 2×10^{17} incident photons/cm^2sec). However, for different cadmium sulfide crystals, LIMA curves differ in shape and amplitude as shown by comparison of curves (1) and (3) in figure 2. (This excitation density for both curves was the same.) The infrared parts of curve (1) obtained after changing to a different detector (from S-20 to S-1 response photomultiplier for curve (4) to a PbS-cell for curve (5)) are not normalized to equal sensitivity. Two LIMA maxima are observed at about 800 nanometers and 1400 nanometers. These maxima suggest a close correspondence to well-known photoconductivity and photoluminescence quenching maxima.

As shown in figure 3(a), the magnitude of LIMA varies strongly with excitation (laser) intensity. In the bleaching region (650 nm), it depends linearly upon laser intensity whereas in the increased absorption region (740 nm) the dependence is sublinear. This difference indicates a transition between monomolecular and bimolecular recombination kinetics.

LIMA increases with decreasing chopping frequency around 50 hertz (fig. 3(b)) and this result indicates time constants for electron redistribution between 10 and 100 milliseconds.

The observed properties of LIMA suggest that the laser excitation causes a redistribution of electrons over the defect levels in the band gap. Electrons are pumped from levels below the Fermi level into levels closer to the conduction band by way of transitions over this band. Thereby, short-wavelength optical absorption (labeled (1) in fig. 4) is reduced and long-wavelength absorption (labeled (2), (3), and (4)) is increased, as observed. (See fig. 2.) A comparison between LIMA-curves and the spectral distribution of photoconductivity should permit one to separate competing transitions (2) (or (3)) and (4).

CONCLUDING REMARKS

Phenomenologically, the LIMA effect is the broad-band modulation of very weak bulk absorption in a solid. It can be produced by modulated intense light of a wavelength which is weakly absorbed. A more detailed analysis of LIMA should offer a new and valuable tool for direct investigation of the redistribution of electrons over defect levels and for improving the understanding of defect structure of solids.

Langley Research Center,
National Aeronautics and Space Administration,
Langley Station, Hampton, Va., February 19, 1968,
129-03-15-04-23.

REFERENCES

1. Park, Kwangjai: Excited State of the F Center in KI. Phys. Rev., Second ser., vol. 140, no. 5A, Nov. 29, 1965, pp. A1735-A1741.

2. Fröhlich, Dietmar, and Mahr, Herbert: Decay of Laser-Induced Excitations of F Centers. Phys. Rev., Second ser., vol. 141, no. 2, Jan. 1966, pp. 692-695.

3. Maeda, Kieji; and Kasami, Akinobu: Behavior of Holes and Electrons in CdS Revealed by Laser Excitation. II-IV Semiconducting Compounds – 1967 International Conference, D. G. Thomas, ed., W. A. Benjamin, Inc., 1967, pp. 1323-1334.

4. Chiarotti, G.; and Grassano, U. M.: The Excited States of the F Center Investigated by Means of Modulated Optical Absorption. Nuovo Cimento, ser. 10, vol. XLVI B, no. 1, Nov. 11, 1966, pp. 78-92.

5. Hopfield, J. J.; Worlock, J. M.; and Park, Kwangjai: Two-Quantum Absorption Spectrum of KI. Phys. Rev. Letters, vol. 11, no. 9, Nov. 1, 1963, pp. 414-417.

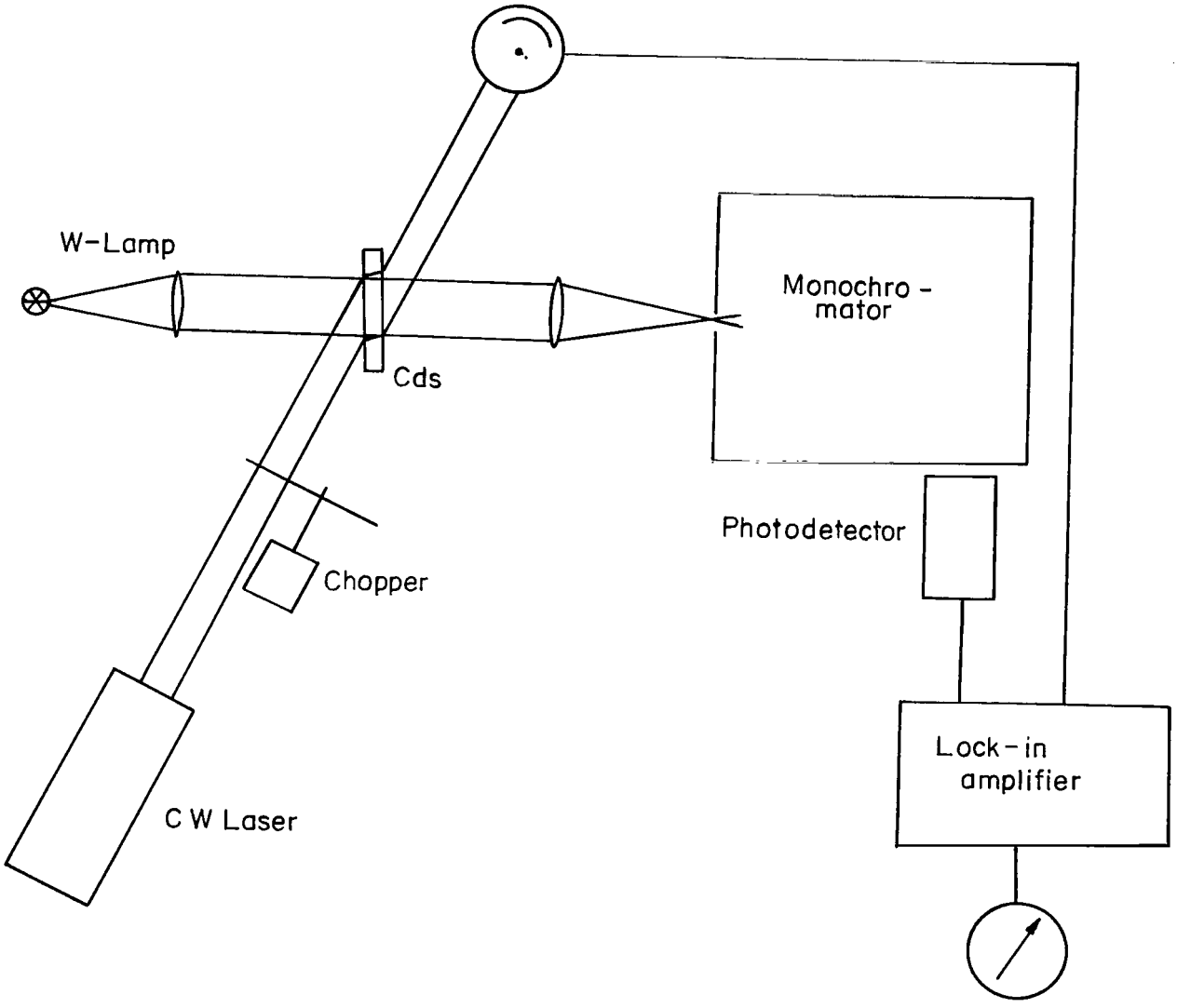


Figure 1.- Experimental setup.

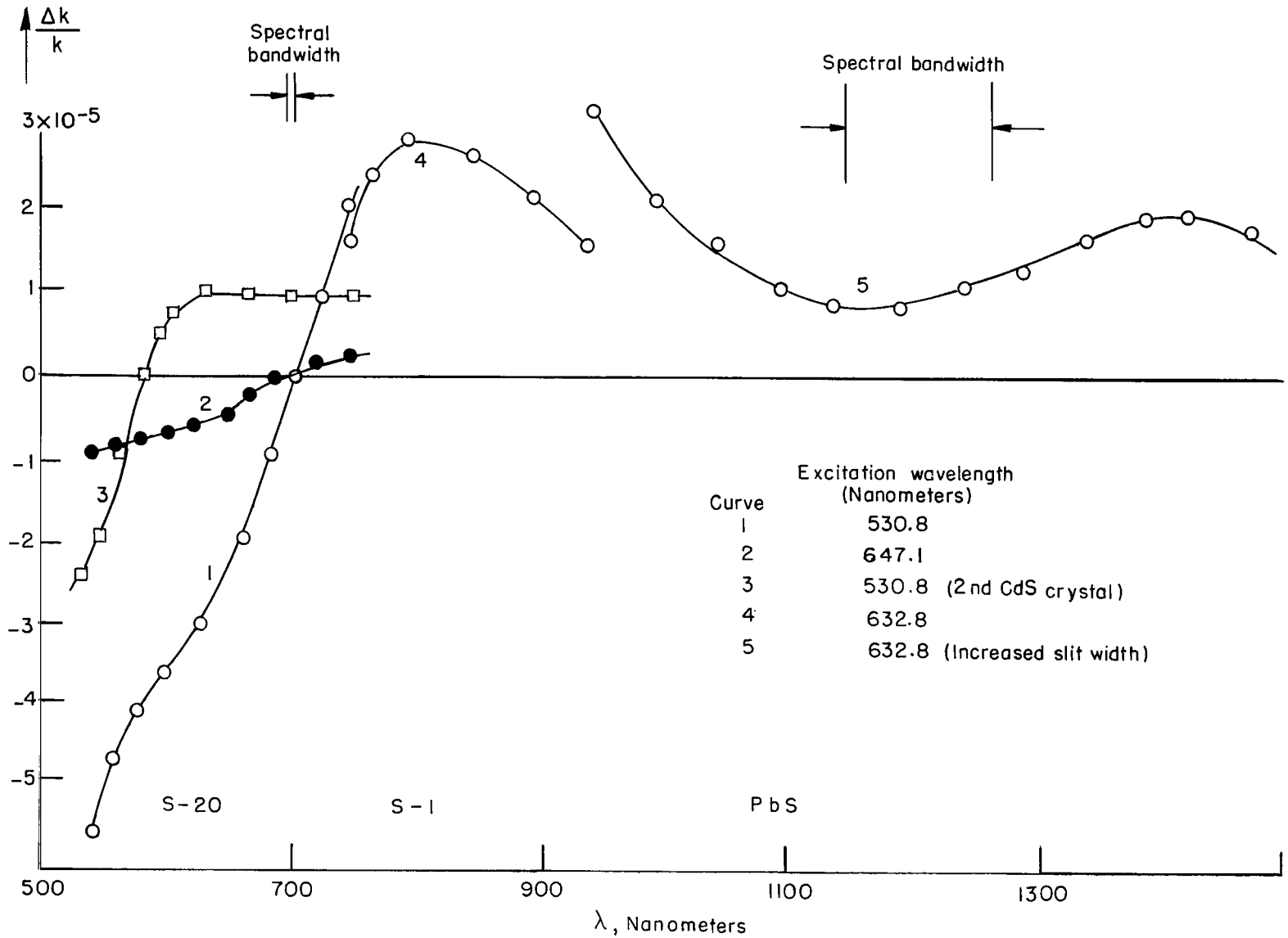
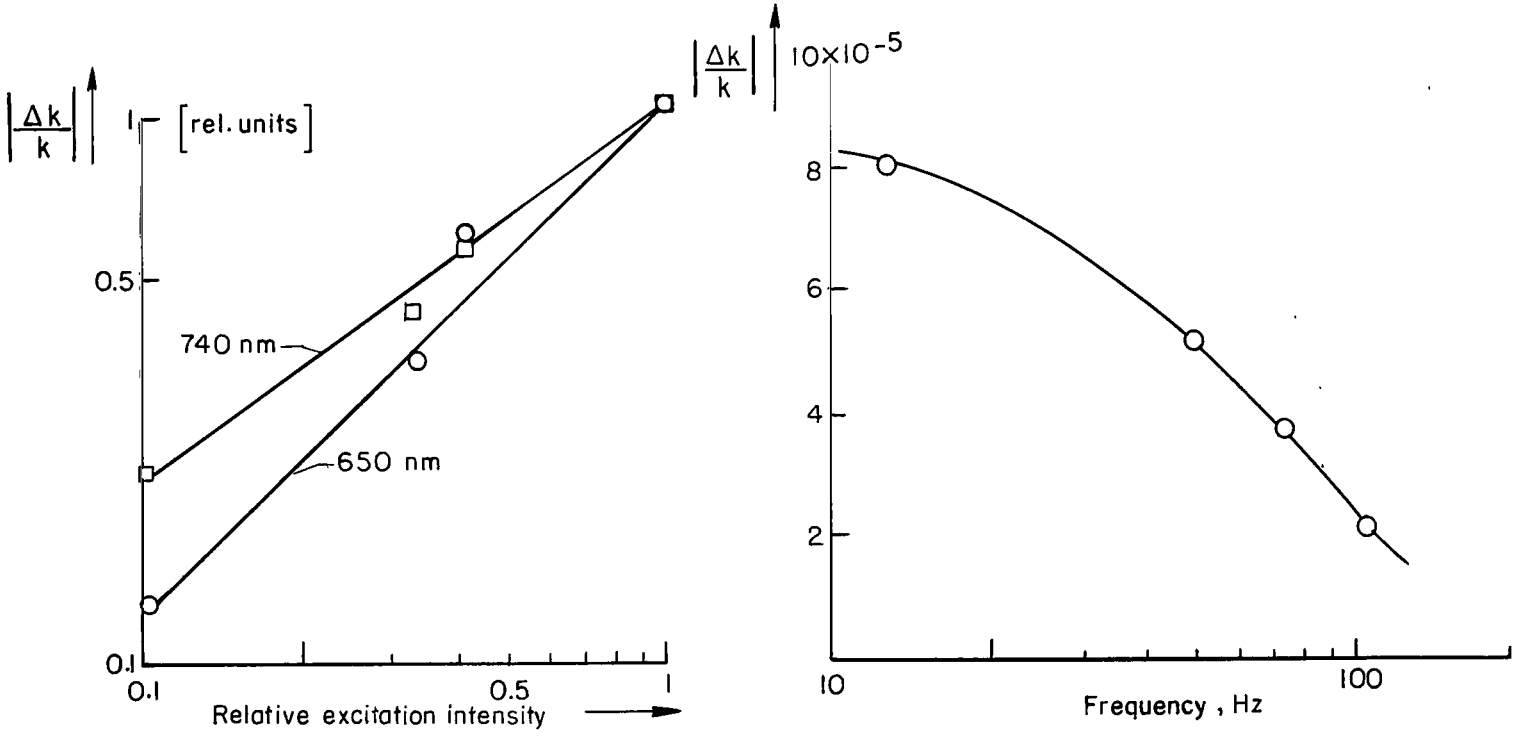


Figure 2.- Relative optical absorption as a function of wavelength.



(a) Variation with excitation intensity.

(b) Variation with frequency.

Figure 3.- Magnitude of relative change in absorption.

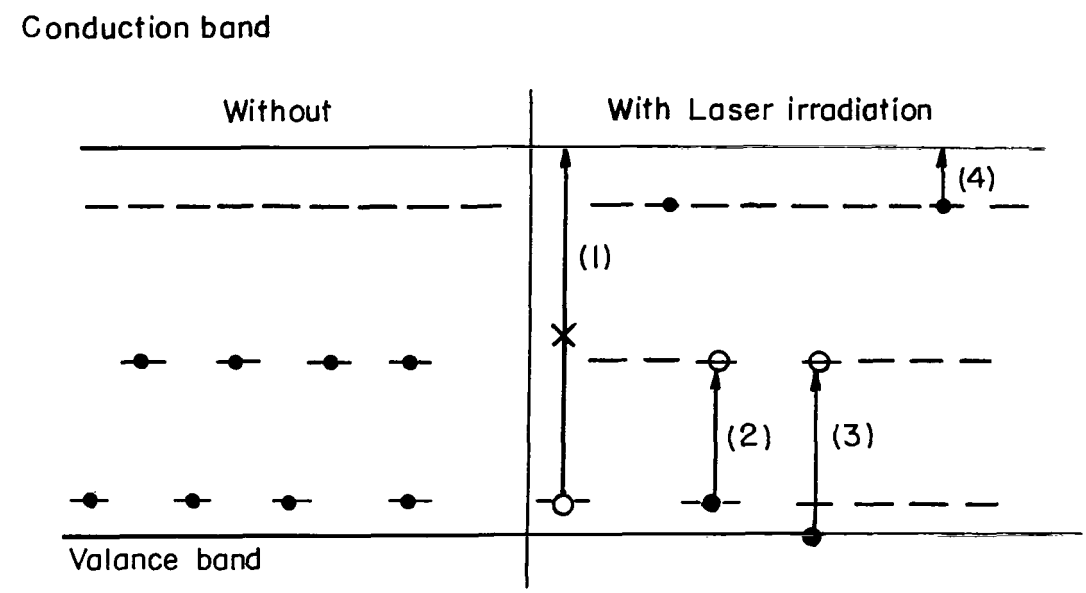


Figure 4.- Band model.

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